RL-TR-91-50
Final Technical Report
May 1991

EFFECTS OF DEPOLARIZATION ON THE MICROWAVE PROPERTIES OF COMPOSITES

Northeastern University

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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY (Leave Blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	May 1991	Final Jul 89 - Dec 89

4. TITLE AND SUBTITLE
EFFECTS OF DEPOLARIZATION ON THE MICROWAVE PROPERTIES OF COMPOSITES

5. FUNDING NUMBERS
C - F30602-88-D-0028
Task E-9-7117
PE - 61102F
PR - 2305
TA - J4
WU - P7

6. AUTHOR(S)
C. Vittoria

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Northeastern University
Boston MA 02159

8. PERFORMING ORGANIZATION REPORT NUMBER
N/A

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR
Bolling AFB DC 20332-6445

Rome Laboratory (EECT)
Hanscom AFB MA 01731-5000

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
RL-TR-91-50

11. SUPPLEMENTARY NOTES
Rome Laboratory Project Engineer: Mark K. Hinders, Captain, USAF/EECT/(617) 377-4265

Prime Contractor: University of Dayton, Graduate School of Engineering, (See reverse)

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)
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NOTE: Rome Laboratory/RL (formerly Rome Air Development Center/RADC)

14. SUBJECT TERMS
Electromagnetics, Microwave Properties, Depolarization

15. NUMBER OF PAGES
28

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

Block 11 (Cont'd)

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1 BACKGROUND

The microwave properties of composite materials are of great interest to many, since it is possible to control or design the constituents of a composite in order to obtain a desireable property of the composite. There are many physical parameters that can effect the microwave properties of a composite and they are: binder material, particle size, shape, electrical and magnetic properties of the particle and binder, density of particles, randomness of particle distribution, etc... In this report we will concentrate our attention on the effect of particle shape on the microwave properties of a composite.
In particular we will consider metallic as well as insulating particles distributed randomly in a known binder material. The effect of the particle shape in composite is to induce electrical and magnetic depolarization fields within the particle itself as well as beyond the particle. Depolarization fields are induced within magnetic particles, if the magnetization is uniform throughout the particle - single domain. In multi-domain configurations the depolarizing field within the magnetic particle is reduced and may be assumed to be zero for practical purposes. The density of magnetic domains nucleated in a particle depends on the size and shape of the particle. Similar observations can be made for the electric depolarization field, if the particle is ferro-electric, for example. However, for most applications scalar dielectric constant materials are used in composites. Hence, we will assume scalar dielectrics in this report. In such cases the shape of the particle plays a major

role in the magnitude of the depolarizing electric field within the particle. For dilute concentrations of particles in a binder it is meaningful to confine depolarizing fields within the particle. This assumption breaks down, if the density of particles increases beyond 25% (by volume) of the composite material. We will now consider firstly metallic iron alloy particles with dimensions of the order of 20μm imbedded in castolite. For the insulating particles we will consider ferrite particles imbedded in a binder material characterized by high dielectric constant.

2 METALLIC PARTICLES

Composite materials were fabricated in which iron alloy particles were imbedded into castolite binder. Castolite is a dielectric material with a corresponding value of dielectric constant of 2.59-j0.05 over the frequency range of 1 to 20 Ghz. The iron alloy particles were sifted through a 20μm grid mesh. Thus, some particles were greater and others less than 20μm in size. The shape of the particles was unknown. The samples were designated as one set being greater and the other less than 20μm. Toroids of 90% castolite and 10% particles (by volume) were fabricated in the microwave laboratory for the purpose of measuring ϵ (dielectric constant) and μ (permeability constant) of the composite. A network analyzer was used to measure ϵ and μ from 0.5 to 18.6 GHz.

For the composite (characterized by particles greater than 20μm in size) the average dielectric constant was measured to be

$$\epsilon_{av} = 5.40 - j0.080 \qquad (>20\mu m) \quad ,$$

whereas for the other case

$$\epsilon_{av} = 4.25 - j0.075 \qquad (<20\mu m)$$

The value of ϵ remained fairly constant from 0.5 to 18.6 GHz. In analyzing the data we made use of the well known formula of Maxwell-Garnett[1]

$$\epsilon_{av} = \epsilon_b + \frac{F\,(\epsilon - \epsilon_b)}{(1 - F)(1 + L(\epsilon - \epsilon_b)) + F} \qquad (1)$$

where

ϵ = dielectric constant of particle

ϵ_b = dielectric constant of binder material

F = fractional volume of particle loading

L = depolarizing factor

For a conductive medium ϵ may be related to the conductivity by the relation

$$\epsilon = \sigma/j\omega$$

where

$$\sigma = \frac{\sigma_o}{1 - j\omega\tau}$$

τ = relaxation time between collisions.

σ_o = DC conductivity.

If the particle is conductive, eq. (1) reduces to approximately

$$\epsilon_{av} \approx \frac{F}{(1 - F)\,L} + \epsilon_b \qquad (2)$$

Re-write above equation as follows, since F = 0.10 for our composites.

$$\frac{\epsilon_{av}}{\epsilon_b} \simeq 1 + \frac{1}{9L\epsilon_b} \qquad (3)$$

Figure (1) shows a plot of ϵ_{av}/ϵ_b versus $\frac{1}{\epsilon_b L}$. We see that for our case that $\ell = \epsilon_b L = \frac{1}{9}$ for the measured value of $\epsilon_{av}/\epsilon_b \simeq 2$

corresponding the composite containing particles greater than $2\mu m$ and $\ell = \frac{1}{5.5}$ for the other composite ($< 20\mu m$).

The fact that the depolarizing factor depends on the size of the particles implies that the shape of these particles are dependent on the size of the particles. The particles tend to be more like disc types for particles' diameter bigger than $20\mu m$.
The same depolarizing factor must also be effective in describing the magnetic properties of the particle. Hence, if we were to apply equation (1) to permeable materials, it would be written as follows

$$\mu_{av} = 1 + \frac{F(\mu - 1)}{(1 - F)(1 + \ell(\mu - 1)) + F} \qquad (4)$$

where μ_b = permeability constant of binder = 1

μ = permeability constant of particle.

The permeability of the particle may be approximated as follows for relative large size samples

$$\mu = 1 + \frac{\chi_o (1 + jx/2Q)}{1 - x^2 + jx/Q} \quad , \qquad (5)$$

where $\chi_o = 4\pi M/H_A$

$Q = f_o/\Delta f$

$x = f/f_o$

$4\pi M$ = Saturation Magnetization.

H_A = Uniaxial magnetic anisotropy field.

f_o = ferromagnetic resonant frequency = γH_A.

f = Operating frequency.

Δf = ferromagnetic resonance linewidth $\triangleq \gamma 2\pi M$

$\gamma \triangleq 2.8$MHz/Oe.

The factor Q is a measure of the material losses at microwave frequencies. It is important to note that the above equation is only appropriate to particles in which multi-domains are nucleated in a fairly high density. Particles in the order of 20μm contain a very high density of multi-domains. It is estimated that one needs to fabricate particles in the order of 1/4 μm in order to energetically favor single domain excitations instead of multi-domains. For single domain excitations equation (5) is inappropriate, because eq. 5 does <u>not</u> take into account of demagnetizing fields [2] or depolarizing fields. There are other complications which the reader should be aware of in the use of equation (5). In a conductive medium eddy currents effects tend to mask or screen the external electromagnetic fields. As such there are diamagnetic[3] contributions to μ. In addition the electromagnetic wave is attenuated in the metallic particle. The attenuation is further enhanced by the exchange conductivity effects[4]. The result of this is to slightly shift f_o from the simple relation γH_A and increase Δf. Estimates of these shifts can be calculated[4] and incorporated in the expression for μ in eq. (5) by slightly modifying f_o and Δf.

Let's calculate μ for a single particle explicitly assuming the following

$4\pi M = 21{,}500$ G

H_A = 500 Oe

Δf = 30 GHz

f_o = 1.5 GHz

The values for these parameters are assumed on the basis that we are dealing with iron rich alloy particles and the cubic magnetic anisotropy field is in the order of 500 Oe [5] and $4\pi M$ is close to that of pure iron. The calculational procedure is as follows: The above parameters together with an assumed frequency value are substituted into eq. (5) and μ is calculated for the single particle. In order to calculate μ_{av} of the composite the value of μ of the particle is substituted into eq. 4 assuming F = 0.10. In figs. (2) and (3) μ_{av} of the composite are calculated for $\lambda = \frac{1}{9}$ and $\frac{1}{5.5}$, respectively.

The most prominent feature of the calculations is that μ'_{av} is constant, where μ'_{av} is the real part of μ_{av}, which is in contrast to measured values of μ'_{av}. In fig. (4) we have plotted μ_{av} calculated from

$$\mu_{av} \simeq 1 + F\,(\mu - 1) \qquad (6)$$

In equation (6) we make the assumption that there are no depolarizing factors in multi-domain excitations in the magnetic particles. In fig. (4) both calculated and measured μ'_{av} scale inversely with frequency with approximately the same slope. Qquantitatively, it appears that inserting depolarizing factors in the expression for μ'_{av} improves the fit to the experimental observation. However, the agreement is only good at low frequencies and, furthermore, calcualted μ'_{av} is constant with frequency. Omitting depolarizing factors eq. (6) predicts a scaling of μ'_{av} with frequency, but quantitatively a factor of 1.7 smaller. We believe that it is important to predict the frequency dependence of μ'_{av}, since there are other contributions which can account for the quantitative difference. We will discuss these contributions at the end of this section.

Also calculated $\mu"_{av}$ is in reasonable agreement with measured $\mu"_{av}$ considering the measured values of $\mu"_{av}$ are relatively small compared to one. Network analyzer techniques are not amenable to measuring small values of $\mu"_{av}$.

The difference between the measured and calculated values of $\mu"_{av}$ may be accounted by three equally important factors: (1) Since the particles are conductive, only material within the skin depth distance from the particle surface is magnetically active. Effectively, this means that the actual filling factor F is smaller than 0.1 we have assumed above in equation (6) toroids. (2) Thus, if we were to choose for example F = 0.04, better agreement can be obtained between measured and calculated values of μ'_{av}. However, even assuming F = 0.04 there is still discrepancy in $\mu"_{av}$. The measured $\mu"_{av}$ is a factor of two bigger than the calculated value. This difference may be accounted for by additional eddy current losses found in conductive materials. This extra loss manifests itself in the measurement of $\mu"_{av}$. (3) Finally, equation (5) is indeed an approximate equation. Accurate expression for μ for specific multi-domain configuations may be obtained in reference (6). The parameter values for Δf and f_o were chosen in a manner to make the resonance curve highly asymmetrical. Equation (5) applies only to resonance curves which are symmetrical with respect to frequencies below above f_o. Nevertheless, we feel that reasonable agreement between measured and calculated values of μ_{av} was obtained.

3 INSULATING PARTICLES

For insulating particles one may ignore eddy current[3] and exchange conductivity[4] effects. With these omissions eq. (5) should be more relevant to insulating particles. However, the expression for ϵ needs to be modified to the following

$$\epsilon = \epsilon_o \left[1 + \omega^2 p \Big/ \left(\left(\omega^2{}_o - \omega^2 \right) + j \frac{\omega}{\tau} \right) \right]$$

ϵ_o = dielectric constant of free space

ω_p = plasma frequency

ω_o = electronic resonance

Data was provided to us on a composite material which consisted of the following constituents: Ni-zn ferrite particles[7] which exhibited saturation magnetization of ($4\pi M$) 4200 G, H = 3600 Oe, F = 0.62, ϵ = (20.9 - 0.48 f) - j(0.7 - 0.005f). The dielectric constant of the binder, ϵ_b, was 2.8. In fig. (5) calculated and measured values of ϵ_{av} have been plotted as a function of frequency from 2 to 18 GHz. Calculated values were obtained using equation (1) and assuming the following parameter values: ϵ_b = 2.8, F = 0.62 and

$$L = \frac{1}{3\epsilon_b} .$$

We now apply equation (5) and (6) to calculate μ_{av} of the composite assuming above magnetic parameters. In fig. (6) the permeability of the single particle is plotted as a function of frequency. The comparison between calculated and measured μ is reasonable considering that only two adjustable parameters are used in eq. (5). Improved comparison may be obtained, if Δf is lowered with respect to the assumed value of 5.88 GHz. However reduction of Δf is not consistent with maintaining $4\pi M$ constant, since $\Delta f = \gamma 2\pi M$. In fig. (7) we calculated μ_{av} using eq. (6) together with eq. (5). Again only two adjustable parameters are assumed, H_A and $4\pi M$, which also give $f_o = \gamma H_A$ = 10.08 GHz and $\Delta f = 2\pi M$ = 5.88 GHz. In fig. 7 the agreement between calculated and measured μ_{av} is reasonable if we were to lower f_o by as much as 2.2 GHz in eq. 5. There is no physical reason to do so. Further examination of the material preparation of the ferrites it has been revealed that the ferrites were sintered at high temperature

prior to the fabrication of the composite. It is well known that sintering[2,5] processes in ferrites can significantly alter the values of H_A, for example. Hence, we conclude that the value of H_A was lowered by roughly 750 Oe by the process of sintering.

4 CONCLUSIONS

1. Depolarizing factors are important in the calculation of the average value of the dielectric constant in composite materials consisting of dielectric particles. For metallic particles propagation effects must be considered especially for composites made up of relative large size particles in comparison to the skin depth.

2. For large size magnetic particles imbedded in composites the depolarizing factor may be omitted in the calculation of the average value of the permeability of composite materials.The depolarizing factor may be omitted, since multi-domains are nucleated in large size magnetic particles. The depolarizing field averages out to zero for multi-domain configuration: Our equation (5) is a reasonable estimate of μ in the regime of multi-domain excitations. However, caution must be exercised in the use of equation (5), if the domain density is small and close to unity. Exact calculations must be invoked, if improved estimates of μ are desireable for low domain densities. For single domain excitations as in very small magnetic particles the reader is referred to reference (2) for the calcualtion of μ. In this limit one then must also include depolarizing factors in the calculation of μ and μ_{av} (eqs. (5) and (4)).

3. The above two conclusions are applicable for dilute concentration of particles in a composite material. For large concentrations of particles in a composite, particle

to particle interactions must be included. The interactions may be electrostatic or magnetostatic in nature. This is still an outstanding theoretical problem which is of current interest.

4. We have proposed a two parameter model in the calculation of both μ and μ_{av}, namely H_A and $4\pi M$. f_o and Δf scale as H_A and M, respectively. HOwever, microwave data on μ and μ_{av} seems to suggest at least one more parameter is needed to explain the data completely. A simple way to modify our present analysis is to allow Δf to be a variable, when substituted in eq. (5). As a future guide one may assume a maximum value of $\Delta f \simeq \gamma\, 2\pi M$ and a minimum value in the order of the intrinsic linewidth of the single particle.

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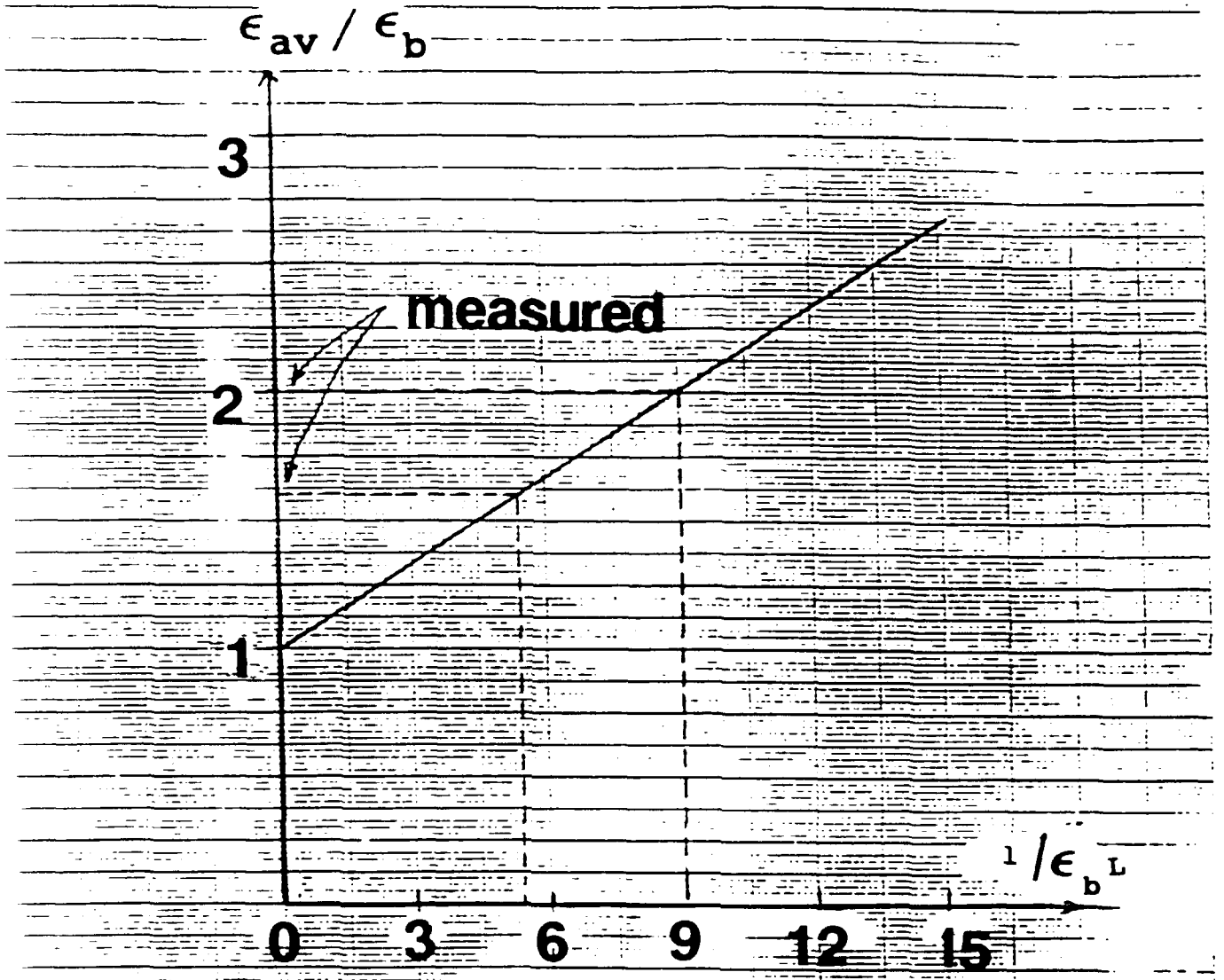

Fig.1

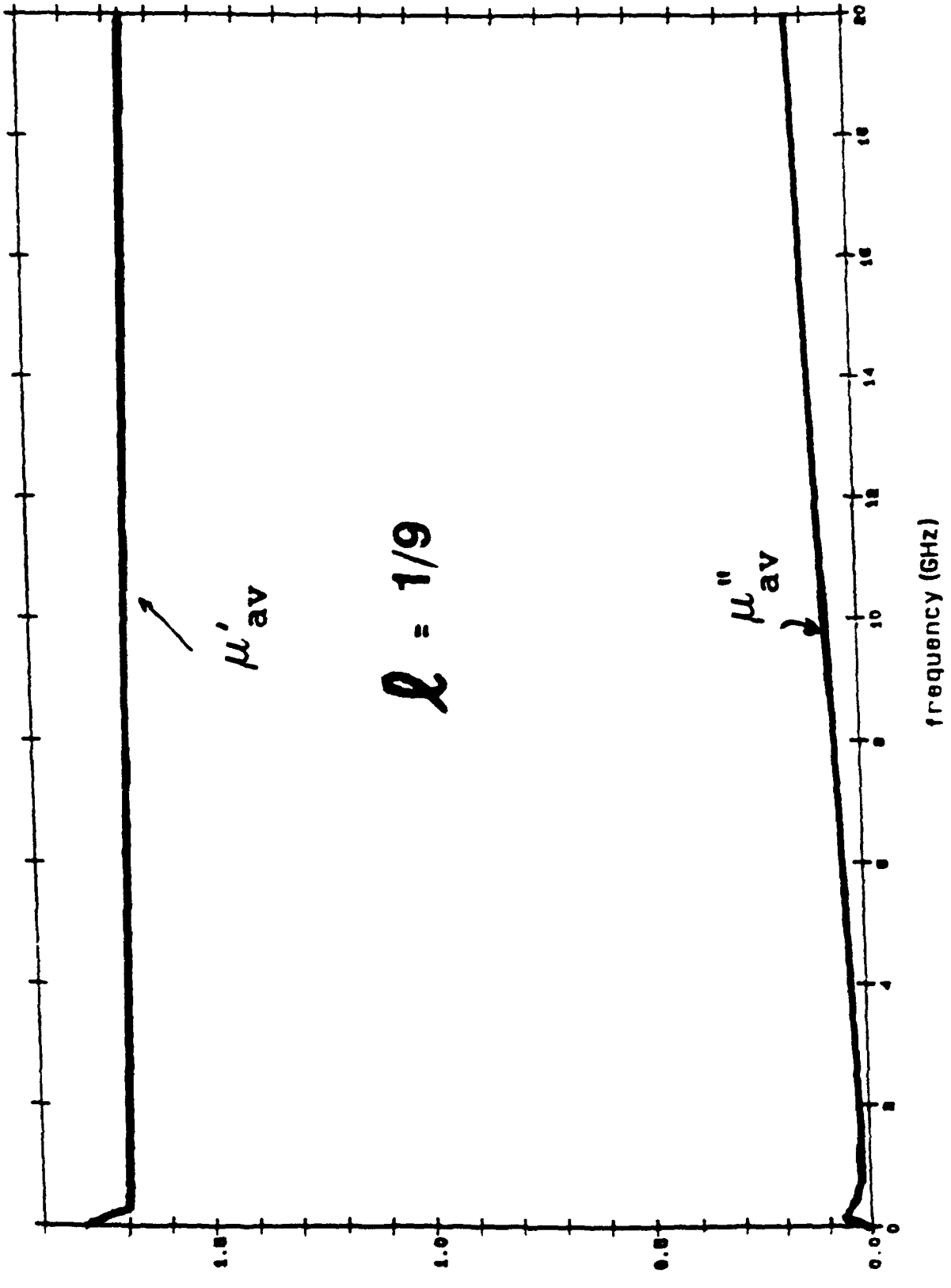

Fig.2

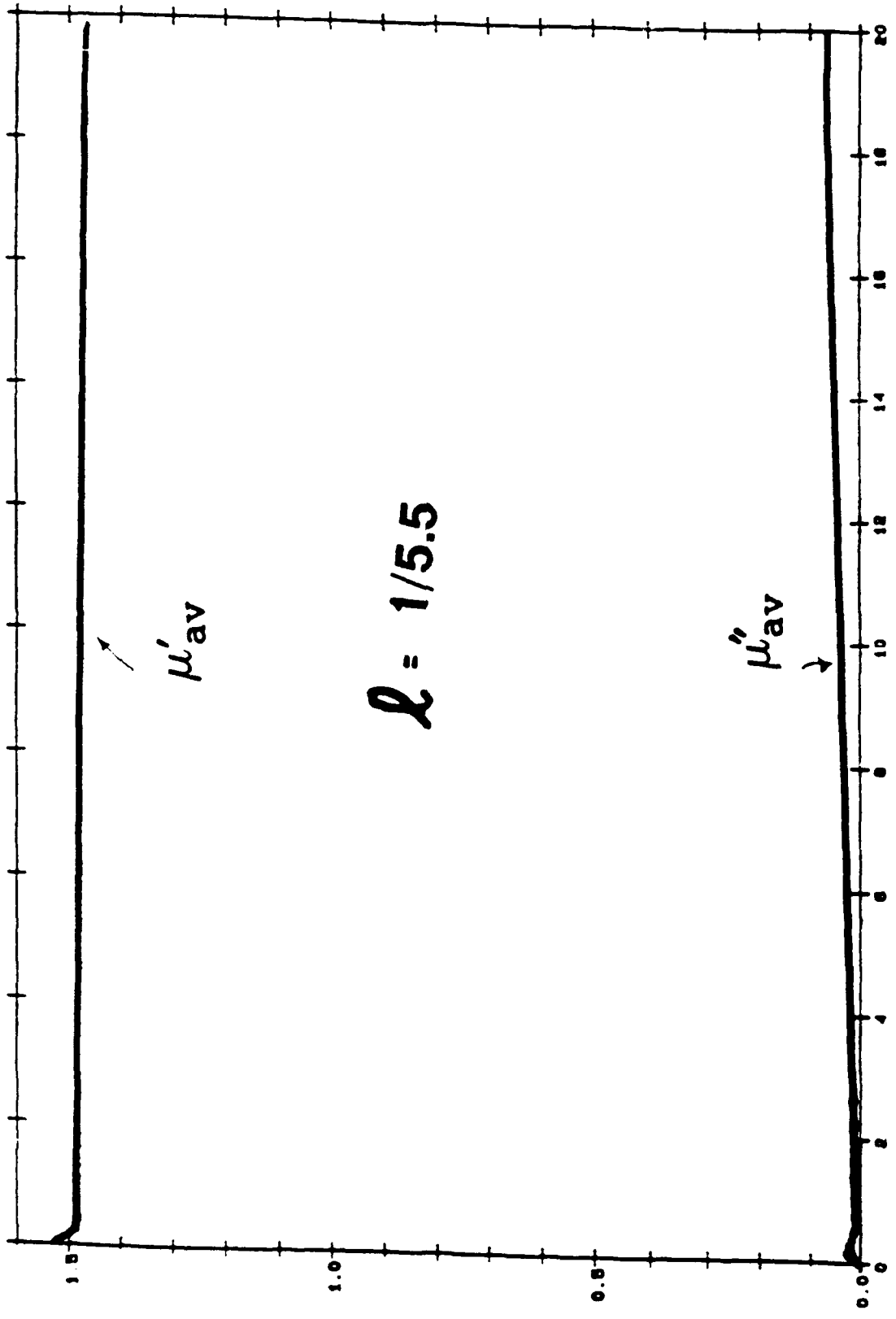

Fig.3

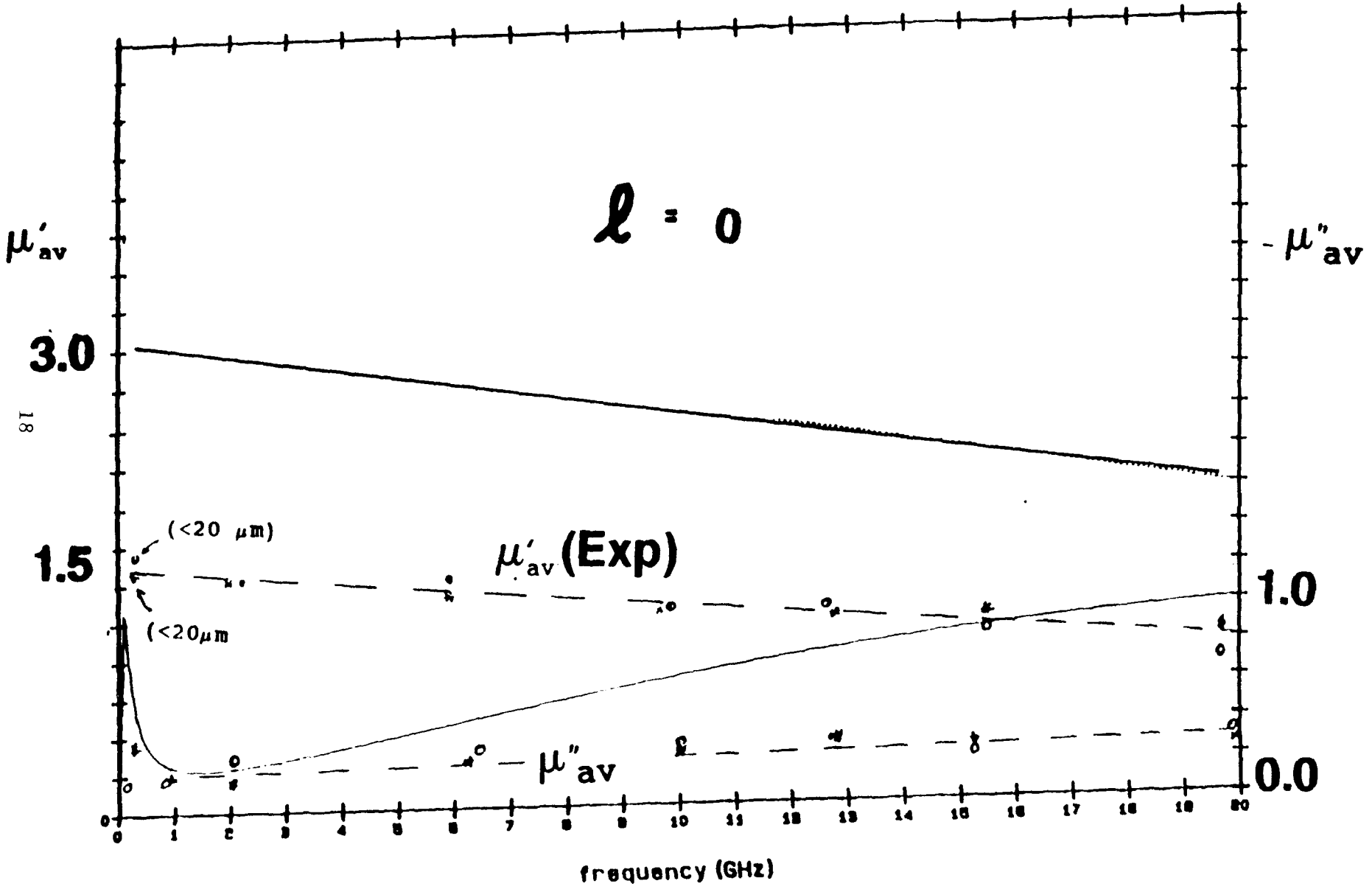
ℓ = 0
μ'_{av}
$-\mu''_{av}$
3.0
1.5
1.0
0.0
(<20 μm)
(<20μm
μ'_{av}(Exp)
μ''_{av}
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20
frequency (GHz)

Fig.4

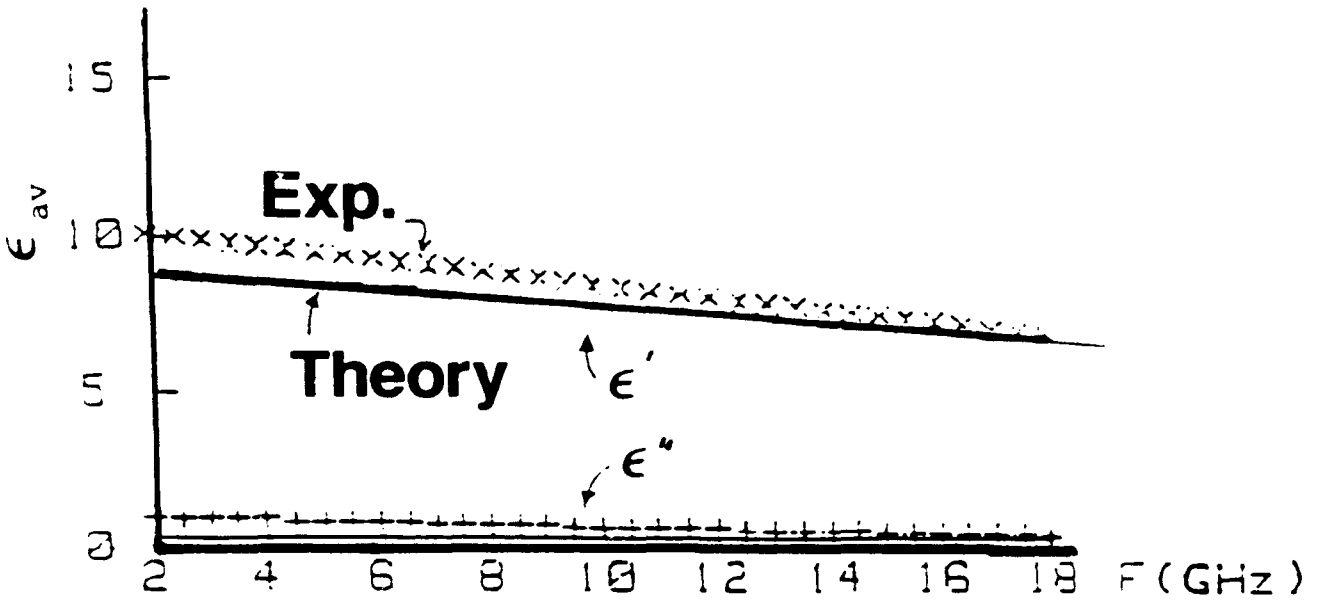
15
10
5
0
ϵ_{av}
Exp.
Theory
ϵ'
ϵ''
2
4
6
8
10
12
14
16
18
F(GHz)

Fig.5

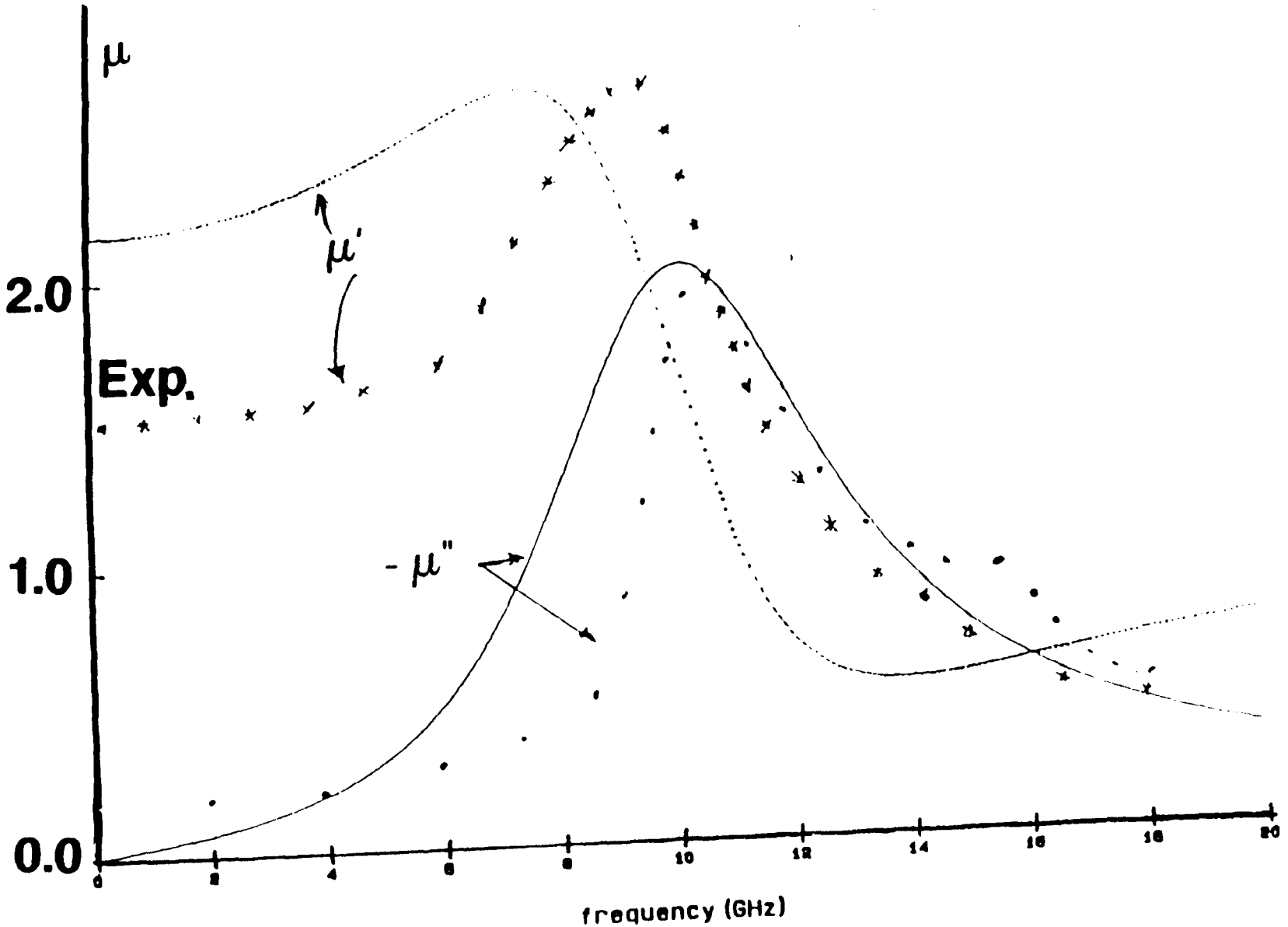

Fig.6

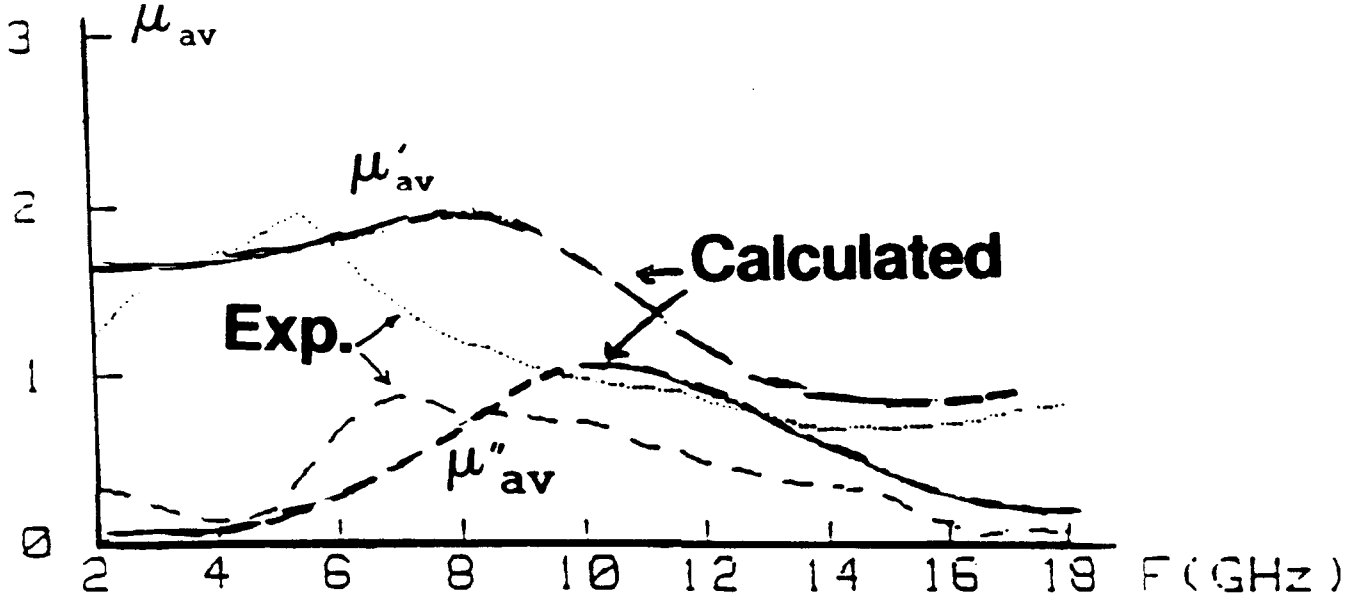

Fig. 1

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